

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:) Group Art Unit: 1797
Thiagarajan et al.) Confirmation No: 4924
)
Serial No. 10/532,843) Examiner: Bullock
Filed: December 19, 2005)
)
For: METHOD FOR) Attorney Docket: 1-17016
 CATALYTICALLY)
 DEHYDRATING)
 HYDROCARBONS)

June 7, 2010

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

BRIEF ON APPEAL

Honorable Sir:

This brief is in furtherance of the Notice of Appeal, which was timely filed in connection with the above-captioned application on April 5, 2010. This Brief is being filed under the provisions of 37 CFR §41.37 and its related requirements. The fees required under 37 CFR 1.17(F) are submitted herewith. As this brief was due on Saturday, June 5, 2010, it is submitted that the submission today, Monday, June 7, 2010, is timely under the provisions of 37 CFR §1.7.

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1. Real Party in Interest

The real party in interest in Uhde GmbH, which is a subsidiary of Thyssen-Krupp
GmbH.

2. Related Appeals and Interferences

There is no known appeal or interference which will directly affect, or be directly affected by, or have a bearing on, the Board's decision in this Appeal.

3. Status of Claims

On April 5, 2010, applicant submitted a Notice of Appeal in connection with the subject application, appealing the final rejection of claims 10-21.

The status of each of the claims is as follows:

1. Claims cancelled: None;
2. Claims withdrawn from consideration but not cancelled: None;
3. Claims pending: 10-21;
4. Claims allowed: None;
5. Claims rejected: 10-21.

The claims on appeal are 10-21. A copy of the claims on file is submitted in the attached Claims Appendix.

4. Status of Amendments

No amendment was filed subsequent to the final rejection of the application by the Office Action of January 5, 2010.

5. Summary of Claimed Subject Matter

Claim 10

The present invention, as defined by independent claim 10, defines a process for the production of unsaturated hydrocarbons, with the following layout. In a first step hydrocarbon, which may have a water vapor content but which is essentially free from oxygen, penetrates in a continuous stream through a first catalyst bed, the latter exhibiting the standard dehydration conditions. Subsequently the reaction mixture obtained in the first step is mixed with liquid phase water and water vapor as well as with an oxygen-bearing gas. Then the reaction mixture is fed in a continuous stream to a further catalyst bed as a second step in which hydrogen oxidation and further dehydration of the hydrocarbons take place.

Support for the present invention as defined in claim 10, can be found as follows:

Support for a hydrocarbon, can be found, at least, on page 4, line 1.

Support of the hydrocarbon possibly having a water vapor content but which is essentially free from oxygen, can be found , at least, on page 4, lines 1 and 2.

Support for the step of the hydrocarbon penetrating in a continuous stream through a first catalyst bed can be found, at least, on page 4, line 3.

Support for the first catalyst bed exhibiting the standard dehydration conditions can be found, at least, on page 4, lines 3 and 4.

Support for the reaction mixture obtained in the first step being mixed with liquid phase water can be found, at least, on page 4, line 7 and further described on page 4, lines 16-21.

Support for the reaction mixture obtained in the first step being mixed with water vapor can be found, at least, on page 4, line 7, and further described on page 4, lines 16-21.

Support for the reaction mixture obtained in the first step being mixed with an oxygen-bearing gas can be found, at least, on page 4, line 7.

Support for feeding the reaction mixture in a continuous stream to a further catalyst bed can be found, at least, on page 4, line 8.

Support for hydrogen oxidation and further dehydration of the hydrocarbons taking place in the further catalyst bed can be found, at least, on page 4, lines 8-10.

Claim 21

The present invention, as defined in independent claim 21, defines a process for the production of unsaturated hydrocarbons. In a first step a hydrocarbon, which may have a water vapor content but which is essentially free from oxygen, penetrates in a continuous stream through a first catalyst bed, the latter exhibiting the standard dehydration conditions. Subsequently the reaction mixture obtained in the first step is mixed with liquid phase water and water vapor as well as with an oxygen-bearing gas. The water and water vapor introduction decreases the reaction temperature. Then the reaction mixture is fed in a continuous stream to a further catalyst bed as a second step in which hydrogen oxidation and further dehydration of the hydrocarbons take place.

Support for the present invention as defined in claim 21, can be found as follows:

Support for a hydrocarbon, can be found, at least, on page 4, line 1.

Support of the hydrocarbon possibly having a water vapor content but which is essentially free from oxygen, can be found , at least, on page 4, lines 1 and 2.

Support for the step of the hydrocarbon penetrating in a continuous stream through a first catalyst bed can be found, at least, on page 4, line 3.

Support for the first catalyst bed exhibiting the standard dehydration conditions can be found, at least, on page 4, lines 3 and 4.

Support for the reaction mixture obtained in the first step being mixed with liquid phase water can be found, at least, on page 4, line 7 and further described on page 4, lines 16-21.

Support for the reaction mixture obtained in the first step being mixed with water vapor can be found, at least, on page 4, line 7, and further described on page 4, lines 16-21.

Support for the reaction mixture obtained in the first step being mixed with an oxygen-bearing gas can be found, at least, on page 4, line 7.

Support for the liquid phase water and the vapor phase water decreasing the reaction temperature can be found, at least, on page 4, lines 17 and 18, and further on page 6, lines 14-16.

Support for feeding the reaction mixture in a continuous stream to a further catalyst bed can be found, at least, on page 4, line 8.

Support for hydrogen oxidation and further dehydration of the hydrocarbons taking place in the further catalyst bed can be found, at least, on page 4, lines 8-10.

6. Grounds for Rejection to be Reviewed on Appeal

On January 5, 2010, the Examiner issued an Office Action in connection with the present application, which rejection was made final. The Examiner maintained his rejection of all of the pending claims. Namely:

Claims 10-21 were rejected under 35 USC 103(a) as being unpatentable over WO 96/33150 in view of US 4,739,123 to Ward. The Examiner states that the WO reference discloses a process for converting alkanes to alkene through a series of process steps. The Examiner acknowledges that the WO reference does not disclose the addition of liquid water to the effluent of step (a). The Examiner states that Ward discloses that the effluent can be cooled by direct or indirect means, in which the effluent is mixed with a low temperature cooling media which may be a gas or liquid.

The Examiner from this opines that it would have been obvious to one skilled in the art to add water and/or water vapor in the direct cooling step of the WO reference as disclosed by Ward.

7. Argument

Rejection of claims 10-20 under 35 USC 103(a) as being unpatentable over WO 96/33150 in view of US 4,739,123 to Ward.

Claims 10-20 stand or fall together and will be argued collectively herein, in particular with regard to independent claim 10.

In the Office Action of January 5, 2010, the Examiner states that the WO reference discloses a process for converting alkanes to alkenes comprising contacting the alkane with a dehydrogenation catalyst under conditions sufficient to produce alkene and hydrogen, contacting the effluent with an oxidation catalyst and oxygen, and contacting the effluent with a dehydrogenation catalyst to convert unreacted alkane to additional quantities of alkene and hydrogen. The Examiner acknowledges that the WO reference fails to disclose adding water to the effluent. The Examiner states that it would have been obvious to modify the WO process with the dehydrogenation effluent of Ward.

The Examiner states that Ward discloses a means for increasing conversion in catalytic dehydrogenization processes employing selective hydrogen combustion. This is achieved by having multibed reaction zones, in which effluent from upstream dehydrogenization beds are cooled, either directly or indirectly, prior to such cooled effluent passing an oxidation bed. The direct cooling process may utilize the oxygen-containing gas or vaporized water.

Applicants respectfully disagree with this conclusion of the Examiner. The invention, as defined in claim 10, does not disclose a method for selecting an optimized

cooling method prior to the oxidative heating (selective hydrogen combustion), either direct or indirect, or as a combination of both, but instead is designed to allow a maximum yield of alkene product.

The optimization of such a means for increasing alkene conversion is only possible through a method that maintains control of the coke formation in the catalyst beds. It is known in the art that the dehydrogenation of alkanes leads to significant coke laydown on the catalyst, which deactivates the catalyst over time. Further, it is known that this coke laydown is dependent on the conversion already achieved and on the gas temperature: too high a temperature leads significant coke formation, too low a temperature yields unacceptably low conversion. It is further known that an additional means of controlling coke formation is through the introduction of steam to the process.

In view of the above, the optimization of product yield over time, requires that the coke laydown over different catalyst beds in the series can be controlled such that all of the catalyst beds operate on the same cycle and maintain substantially equal activity over time.

It should be further noted with regard to Ward, that more than 50 to more than 85% of hydrogen entering each oxidation bed shall be consumed by selective combustion, but Ward provides no suggestions or teaching on such optimum hydrogen conversion, as discussed above, while simultaneously controlling coke laydown on the catalyst beds.

To the contrary, the process of the present invention does provide for such a means of optimization. This is introduced through the cooling by a combination of both liquid water and steam.

The introduction of both liquid and gas phase water allows several improvements. Adjustment of the hydrogen combustion to achieve optimum conversion increase by oxidative reheat, while simultaneously controlling maximum temperature achieved by the oxidative reheat thereby adjusting the temperature profile in the various dehydrogenation beds in series with oxidative reheat, and also simultaneously adjusting the optimum steam to hydrocarbon ratio individually in the various dehydrogenation beds in series, allowing the control of coke formation and thereby controlling the activity of the various catalyst beds over the operational cycle.

It is respectfully submitted that one skilled in the art would not look to Ward to combine with the WO reference, and that even if they were such combination would not yield the present invention. One skilled in the art would not be motivated to use the WO reference in conjunction with Ward, and even if combined, would not yield the present invention as claimed herein. Neither of the references teach the combination of steam and water to optimize the process as is done in the present invention.

Additionally, neither of the applied references, and thus no combination of these references, teach the further optimization obtained in that the oxidation and further dehydrogenation may take place in the same catalyst bed and possibly over the same catalyst, which further optimizes the temperature profile, as the endothermic

dehydrogenation reaction reduces directly the maximum temperature achieved by oxidate reheat, thus further reducing the coke formation.

Therefore, it is submitted that claim 10 is allowable over the art of record and reversal of the Examiner's rejection is respectfully requested. Claims 11-20 depend from claim 10 and are thus allowable based on the allowability of claim 10.

Rejection of claim 21 under 35 USC 103(a) as being unpatentable over WO 96/33150 in view of US 4,739,123 to Ward.

Claim 21 stands alone in this appeal.

Claim 21 is substantially similar to claim 10, and therefore all of the arguments regarding claim 10 apply here.

However, claim 21 adds the additional subject matter of specifying that the introduction of water and water vapor cool the reaction temperature. As noted in the specification, this cooling process is significant in that it increases the efficiency of the system. There is nothing in either of the applied references to suggest this function.

The optimization of such a means for increasing alkene conversion is only possible through a method that maintains control of the coke formation in the catalyst beds. It is known in the art that the dehydrogenation of alkanes leads to significant coke laydown on the catalyst, which deactivates the catalyst over time. Further, it is known that this coke laydown is dependent on the conversion already achieved and on the gas temperature: too high a temperature leads significant coke formation, too low a temperature yields unacceptably low conversion. It is further known that an additional means of controlling coke formation is through the introduction of steam to the process.

In view of the above, the optimization of product yield over time, requires that the coke laydown over different catalyst beds in the series can be controlled such that all of

the catalyst beds operate on the same cycle and maintain substantially equal activity over time.

It should be further noted with regard to Ward, that more than 50 to more than 85% of hydrogen entering each oxidation bed shall be consumed by selective combustion, but Ward provides no suggestions or teaching on such optimum hydrogen conversion, as discussed above, while simultaneously controlling coke laydown on the catalyst beds.

To the contrary, the process of the present invention does provide for such a means of optimization. The combination of cooling by both liquid water and steam allows:

Adjustment of the hydrogen combustion to achieve optimum conversion increase by oxidative reheat, while simultaneously controlling maximum temperature achieved by the oxidative reheat thereby adjusting the temperature profile in the various dehydrogenation beds in series with oxidative reheat, and also simultaneously adjusting the optimum steam to hydrocarbon ratio individually in the various dehydrogenation beds in series, allowing the control of coke formation and thereby controlling the activity of the various catalyst beds over the operational cycle.

It is respectfully submitted that one skilled in the art would not look to Ward to combine with the WO reference, and that even if they were such combination would not yield the present invention. One skilled in the art would not be motivated to use the WO reference in conjunction with Ward, and even if combined, would not yield the present

invention as claimed herein. Neither of the references teach the combination of steam and water to optimize the process as is done in the present invention.

Additionally, neither of the applied references, and thus no combination of these references, teach the further optimization obtained in that the oxidation and further dehydrogenation may take place in the same catalyst bed and possibly over the same catalyst, which further optimizes the temperature profile, as the endothermic dehydrogenation reaction reduces directly the maximum temperature achieved by oxidate reheat, thus further reducing the coke formation.

Therefore, it is submitted that claim 21 is allowable over the art of record and reversal of the Examiner's rejection is respectfully requested.

CONCLUSION

As claims 10 and 21 are patentable for the reasons discussed, and as claims 11-20 depend directly or indirectly from independent claim 10, applicant submits claims 10-21 are patentable. An expeditious determination by the Board to that effect is respectfully requested.

Respectfully submitted,

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CLAIMS APPENDIX

Claim 10 (previously presented) A process for the production of unsaturated hydrocarbons, with the following layout:

- In a first step hydrocarbon, which may have a water vapor content but which is essentially free from oxygen, penetrates in a continuous stream through a first catalyst bed, the latter exhibiting the standard dehydration conditions;
- subsequently the reaction mixture obtained in the first step is mixed with liquid phase water and water vapor as well as with an oxygen-bearing gas;
- and then the reaction mixture is fed in a continuous stream to a further catalyst bed as a second step in which hydrogen oxidation and further dehydration of the hydrocarbons take place.

Claim 11 (previously presented) The process according to claim 10, wherein the first catalyst bed is heated and the heating of the first step is preferably adjusted in such a manner that an essentially isothermal operating mode is obtained.

Claim 12 (previously presented) The process according to claim 10, wherein oxygen-bearing gas is added to the reaction mixture produced in the second step and the reaction mixture thus obtained flows in a continuous stream through a further catalyst bed in at least a third step.

Claim 13 (previously presented) The process according to claim 10, wherein the reaction mixture is cooled in a cooling unit downstream of the second step prior to entering a further catalyst bed in a third process step.

Claim 14 (previously presented) The process according to claim 10, wherein the catalyst bed of the first step uses any standard commercial dehydration catalyst and the second and any further catalyst bed are provided with dehydration catalysts that exhibit not only dehydration activity but also SHC activity.

Claim 15 (previously presented) The process according to claim 11, comprising a catalyst that contains Pt and Sn applied to a carrier element essentially containing aluminate.

Claim 16 (previously presented) The process according to claim 11, comprising a specialist catalyst for water oxidation, hence a catalyst that improves the selectivity compared to that of standard dehydration catalysts when it comes to hydrogen oxidation, the said specialist catalysts being employed in combination with standard dehydration catalysts.

Claim 17 (previously presented) The process according to claim 10, wherein the oxygen-bearing gas is oxygen-rich air.

Claim 18 (previously presented) The process according to claim 10, wherein the quantity of oxygen-bearing gas added in the second and further steps is controlled via the temperature measured at the outlet of the respective upstream catalyst bed or via the outlet temperature of the last catalyst bed.

Claim 19 (previously presented) The process according to claim 10, wherein the hydrocarbon comprises a mixture containing alkanes.

Claim 20 (previously presented) The process according to claim 15, wherein the aluminate comprises zinc aluminate.

Claim 21 (previously presented) A process for the production of unsaturated hydrocarbons, with the following layout:

- in a first step hydrocarbon, which may have a water vapor content but which is essentially free from oxygen, penetrates in a continuous stream through a first catalyst bed, the latter exhibiting the standard dehydration conditions;
- subsequently the reaction mixture obtained in the first step is mixed with liquid phase water and water vapor as well as with an oxygen-bearing gas, wherein the water and water vapor introduction decreases the reaction temperature;
- and then the reaction mixture is fed in a continuous stream to a further catalyst bed as a second step in which hydrogen oxidation and further dehydration of the hydrocarbons take place.

EVIDENCE APPENDIX

none

RELATED PROCEEDINGS APPENDIX

none